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The project, "Spectroscopic Investigations of nonambient systems with Millimeter and Submillimeter Probes," addressed a broad range of scientific topics and technological developments important to this spectral region. For the purposes of this discussion, we have divided the report on research results into three parts: (1) Energy transfer in polyatomic molecules; (2) Molecular collisional processes at very low temperature (<5K), and (3) Development and uses of millimeter and submillimeter technology. Also included is a list of personnel who worked on these projects.

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SPECTROSCOPIC INVESTIGATIONS OF NONAMBIENT SYSTEMS WITH

MILLIMETER AND SUBMILLIMETER PROBES

FINAL REPORT

Frank C. De Lucia

March 15, 1993

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U. S. Army Research Office

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TABLE OF CONTENTS

ABSTRACT OF WORK UNDER ARO CONTRACT DAAL03-89-K-0181

The project, "Spectroscopic Investigations of nonambient systems with Millimeter and Submillimeter Probes," addressed a broad range of scientific topics and technological developments important to this spectral region. For the purposes of this discussion, we have divided the report on research results into three parts: (1) Energy transfer in polyatomic molecules; (2) Molecular collisional processes at very low temperature (< 5 K), and (3) Development and uses of millimeter and submillimeter technology. Also included is a list of personnel who worked on these projects.

I. INTRODUCTION AND BACKGROUND

This report is for the first part of a contract that was subsequently transferred to Ohio State in the context of a new contract. The project addressed the development and use of spectroscopic techniques for the study of molecular systems under nonambient conditions. Emphasis was placed on the use of millimeter and submillimeter spectroscopic methods as diagnostic probes of these environments although many of these techniques are also applicable in other spectral regions. In some cases, the mm/submm spectral region was especially advantageous for the work while in others it was merely convenient because of our experience and equipment base in this part of the spectrum. Much of what is discussed is based upon previous developments in our laboratory. In this Introduction, we will briefly discuss some of the basis for this work; in later sections more of the details and references are provided.

A. The mm/submm Spectral Region

   1. The spectroscopic technique: The mm/submm spectroscopic technique which was used in many of these experiments has been developed over a period of years.[1-3] Briefly, nonlinear harmonic generators which produce the mm/submm radiation are driven by klystrons or traveling wave tube amplifiers[4] in the region around 50 GHz. These microwave sources are in turn referenced to or driven by sources which are controlled by frequency synthesizers, effectively giving a synthesized source which is conveniently and continuously tunable throughout the region between ~ 100 - 1000 GHz. This mm/submm energy is radiated quasi-optically through the atomic and molecular system being studied and detected by an InSb detector operating at 1.5 K or a 0.3 K germanium bolometer. Associated electronics are used for frequency measurement and signal recovery. Systems of this type have proven to be reliable, easy to operate, and reasonably inexpensive. Because of these

attributes, a number of similar systems have now been built at laboratories around the world and have become the standard for a wide range of molecular studies in this spectral region.

2. Characteristics of mm/submm spectroscopy: Much of the work discussed in this report depended upon several important attributes of our technique for spectroscopic studies in the mm/submm spectral region. Among these are:

a) It is a very sensitive technique. Under equilibrium conditions, rotational absorption coefficients increase as $v^2 \rightarrow v^3$ and peak typically in the vicinity of 1000 GHz. Under optimum non-equilibrium conditions, achievable in our pumped experiments, concentrations as small as $100 \, cm^{-3}$ ($\sim 10^{-14}$ Torr partial pressure) can be studied with modest signal averaging. Calculations based on a more aggressive set of assumptions yield concentrations as small as $1 \, cm^{-3}$.

b) Because of the broad tunable coverage in the frequency regime that corresponds to many of the rotational transitions of the small, fundamental species that we wish to study, transitions can be chosen on the basis of their diagnostic value rather than to satisfy coincidence criteria. Furthermore, because of the high resolution, spectral overlap is very rare.

c) It is an absorption technique. Because the transitions that are observed are rotational transitions with accurately known transition moments, the absolute absorption data can be accurately converted into molecular information.

d) Because we directly observe rotational transitions, the method is especially sensitive to rotational nonequilibria. For example, at 300 K and 300 GHz a 5 % change in the population of one of the states involved in a transition produces a 100 % change in the observed line strength.

e) Because this is the frequency regime in which a very large number of optically pumped lasers have been discovered, we can take advantage of the hundreds of man years that have been spent searching for CO_2 laser pump coincidences.

B. Applications

Over a number of years we have used these mm/submm techniques for a wide variety of spectroscopic investigations. In the sections below some of those that are related to the reported work are briefly discussed.

1. _Spectroscopy of small, fundamental molecular species:_ The principal initial application of this spectroscopic technique was its use to develop a basic spectroscopic understanding of small, fundamental molecules in the mm/submm. Particular emphasis was placed on species of atmospheric, astronomical, chemical, and spectroscopic interest. This work has included studies of light asymmetric rotors such as H_2O and H_2S, prototype internal rotors such as HOOH and CH_3OH, light asymmetric rotors with electronic angular momenta such as NO_2, HO_2, and HCO, and a number of unstable species and ions such as CCH, NO^+, and LiH. In addition to the experimental aspects of this work, we have also developed theoretical and computational methods for characterizing to microwave accuracy these spectra over wide spectral ranges. Many (if not most) of these studies were the first to provide spectral characterization of these species to microwave accuracy over large portions of their rotational spectra in the mm/submm. This work is of importance to the reported work because the basic spectroscopic properties (energy levels, transition frequencies, transition moments, etc.) of the molecular species which we studied must first be well understood.

2. _Collisionally cooled spectroscopy at very low temperatures:_ As is discussed in more detail in Section III, we have devoted a significant portion of our effort recently toward the development and exploitation of a collisional cooling technique which provides a simple method for the study of gas phase processes at temperatures below 5 K.[5,6] The scientific motivation for this work has been the desire to study molecular interactions in the regime where $kT \leq hv_r$. To this point we have focused our attention on the study of pressure broadening in the region between 5 K and 1.5 K and on investigations of vibrational and rotational relaxation at 77 K. In this regime we have found that the mechanisms responsible for broadening make a transition from being dominated by classical inelastic collisions at higher temperatures to a lower energy regime in which resonances associated with quasi-bound states become important. The variety and richness of the results that we have obtained have been beyond our expectations.

3. _Collisional energy transfer:_ We have also done considerable work on the internal mechanisms of molecular lasers and the basic collisional energy transfer processes that are the basis of them, especially optically pumped FIR lasers.[7,8] Our emphasis has been on rotational processes both because this degree of freedom corresponds most closely with mm/submm devices and because a number of interesting basic problems exist in rotational energy transfer.

II. EXPERIMENTAL AND THEORETICAL CONSIDERATIONS

Because the thrust of this report is the study of molecular systems under nonambient conditions, it is useful to briefly consider the spectroscopic consequences of significant changes in temperature both on spectroscopic parameters and the physics of the systems being studied.

A. Gas Phase Molecular Interactions as a Function of Temperature

First consider the temperature scaling laws for the interaction of gas phase molecular systems and mm/submm radiation. For the purposes of numerical comparison, we will assume that a system originally at 300 K is cooled to 4 K and that it may be diluted in a collisional cooling gas.

1. _Linewidth:_ In the mm/submm spectral region experiments are typically run under doppler broadened limits. Since doppler broadening is proportional to $T^{1/2}$, linewidths at 4 K are typically an order of magnitude narrower and correspondingly greater resolution and measurement accuracy are possible. In addition, since sensitivity is ordinarily determined by peak, not integrated intensity, this factor under many circumstances also adds an order of magnitude to system sensitivity. We have confirmed this expectation in a number of studies.[5,6,9-13]

2. _Sensitivity:_ Several temperature dependent factors affect molecular absorption coefficients. The rotational absorption coefficient is given by

$$\alpha = [8\pi^2 \nu / 3ch] \cdot |<m|\mu|n>|^2 \cdot [N/\Delta\nu] \cdot [1-e^{-h\nu/kT}] \cdot [1/Q_r]$$

where it has been factored to separate the individual temperature dependent terms. The behavior of $N/\Delta\nu$ depends upon the details of the pressure broadening, but simple theory gives

$$N/\Delta\nu \propto T^{-1/2}$$

Q_r is the rotational partition function which approaches 1 in the low temperature limit. If we consider molecules whose $J = 0 - 1$ transitions f ll at ~ 100 GHz, Q_r is ~ 100 - 1000. The term $[1-e^{-h\nu/kT}]$, which represents the difference between induced absorption and emission varies between unity in the limit $h\nu >> kT$ and ~1/100 for transitions of $\nu \sim 100$ GHz at 300 K. Together these factors represent a four or five order of magnitude gain in absorption coefficient for systems as very low temperature. Other factors are more dependent on the details

of the system. The dilution ratio of the spectroscopic gas in the cooling gas will reduce this gain for stable species, but the reduced pressure broadening parameters associated with zero dipole moment collision partners and the gains in detector sensitivity for low background environments will add to the overall system gains. The latter factor will be especially significant when detector noise is the limiting factor in the system. To give two typical examples, the absorption coefficients of gas phase CO and HCN at 4 K are 25 cm^{-1} and 10000 cm^{-1} respectively. Thus, we conclude that even at very large dilutions of the spectroscopic species in the collisional cooling gas, absorption coefficients will be very large. Again, this expectation has been confirmed in our recent work and molecules with small dilution ratios ($\sim 10^{-4} - 10^{-6}$) are easily observable on an oscilloscope screen in real time.

      3. _Physics_: The spectral complexity of many systems is considerably reduced at low temperature. For example, it has been elegantly demonstrated by many workers using free jet expansions that the complexity of rotational structure in room temperature spectra can be dramatically simplified.[14,15] In addition, rotational collisional processes are of both fundamental and practical importance and many articles and reviews on the subject have been written. In a review article, Brunner and Pritchard[16] compare and contrast 'photon' spectroscopy and 'collisional' spectroscopy. They point out that both spectroscopies are characterized (in principle) by massive amounts of data (mostly redundant) that should be calculable from a relatively small number of 'spectroscopic' constants. However, the nature of 'collisional' spectroscopy is such that it is much more difficult to invert the problem so that fundamental parameters may be recovered from the experimental results. Even if these parameters are given, it is usually a formidable computational task to calculate the effects of these collisions either for the comparison of experiment and theory or for use in other problems. This is because of the participation of the very large number of thermally accessible rotational states in each collision process and because of the increased complexity of molecule-molecule/atom interactions in comparison with molecule-field interactions. The dynamics of the interplay between theory and experiment are further hindered by the nature of the available experimental data. Fundamental parameters are recoverable from most experimental data only via complex (and usually unrealizable) deconvolutions. The origin of this difficulty is very similar to the cause of the theoretical complexity, the large number of thermally populated states and the nature of the molecular interaction. Thus, it is concluded that the very large reduction in the number of thermally populated states at low temperature will make the relation between observables and the fundamental interactions substantially more direct. This is one of the principal motivations for our work. Finally, these very low temperatures significantly enhance the production and lifetime of weakly bound complexes such as van der Waals molecules and also provide additional opportunities for the study of

6

ions and other transient species.

B. System Sensitivity as a Function of Temperature

Over much of the mm/submm spectral region, the sensitivity of our experimental technique is directly proportional to the sensitivity of the detector employed. The cooled detectors used in our work approach being ideal detectors in that their NEP's approach being limited by the fluctuations in the blackbody radiation that is incident on them along with the spectrometer signal. In fact, if one of these detectors is completely broadband and can view the entire 300 K blackbody spectrum, its NEP is limited to about 5×10^{-11} W/Hz$^{1/2}$. In the mm/submm the use of cooled filters that only pass the long wavelength portion of the 300 K blackbody along with the spectrometer signal can significantly improve this, and for a cutoff frequency of ν_m the NEP for an ideal broadband detector is given in the long wavelength limit by NEP $\sim kT\nu_m{}^{1/2}$.[17] For a $\nu_m = 1000$ GHz, this correspond to $\sim 10^{-15}$ W. The 1.5 K InSb detectors that we have used in these systems for many years[2] have NEP's in the range of 10^{-12} - 10^{-13} W/Hz$^{1/2}$ and the 0.3 K ^{3}He detector mentioned above has a NEP of the order 5×10^{-15} W/Hz$^{1/2}$, about 20 times better. It is our experience that this has added a similar amount to our system performance. However, the equation above shows that the construction of a more sensitive broadband detector for the mm/submm spectral region is not possible *if it must view a room temperature experiment.*

III. RESULTS

A. Pressure Broadening below 5 K via Collisional Cooling

1. <u>Experimental results</u>: To date we have investigated the helium pressure broadening characteristics as a function of temperature for a number of species including CO, NO, CH₃F, H_2S, DCl, HDO, OCS, and CH_2F_2 in the temperature region below 5 K.[5,6,9,10,11,12,13,18-20] These measurements have been made with the collisional cooling technique which we have described in the aforementioned papers. The expectation (based on the calculations of sensitivity, linewidth, etc. shown above) that the collisionally cooled technique should be generally applicable has thus been borne out. The only 'failures' that we have encountered have been traceable to injector designs that allowed condensation or freezing for some of the less volatile of the spectroscopic gases.

Our development of the collisional cooling technique was motivated by both the general ideas discussed above as well as a desire to carry out specific experimental investigations. In general, we wanted to be able to experimentally investigate a regime for which $h\nu \le kT$ in the microwave spectral region; thus having molecular systems in analogy to atoms at room temperature, with the energy level spacing large compared to kT. However, only part of the analogy between molecules at very low temperature and atoms under ambient conditions holds because at very low temperatures the collision energies are small or comparable to van der Waals binding energies. Thus, the molecular systems have the additional features of systems that can form quasibound states at very low temperature.[21-23] As a consequence, a rich spectrum of results has been observed that is dependent both on the molecular species and rotational state.

Before showing experimental results, it is worthwhile discussing some of the qualitative features that should be expected. Although it is always dangerous to ascribe too physical a picture to processes that are inherently nonclassical, doing so provides in this case a good mechanism for classifying and understanding the results of our experiments.

In the simplest picture of molecular collisions and their contributions to line broadening, the energy defect (which is a measure of the energy that is transferred between internal and external degrees of freedom) and its relation to kT plays an important role. Whenever this defect becomes too large, the efficiency of the collisions in producing broadening and/or inelastic collisions is reduced; and, in the limit of zero translational energy, this efficiency also becomes zero. However, an additional effect can become important in very low energy collisions. Because of the existence of shallow attractive wells in the intermolecular potential, resonances in both the pressure broadening and inelastic cross sections associated with quasibound states can exist. Open channels with significant strength result from the contributions

9

from rotational levels which lie at low enough energy to interact with the shallow wells.

Thus, in the limit of small collision energy there are two countervailing effects; the reduction in cross section due to the growth in the energy defect and the increase in cross section due to the low energy resonances. Because both of these effects are sensitive functions of the intermolecular potential and the energy level structure of the molecule, a wide variety of functional dependencies of cross section on temperature are observed. These qualitative expectations have been borne out by diagnostic calculations of Palma and Green for the CO - He system[24] and by the experimental results described below.

These ideas lead to the expectation that the pressure broadening cross sections of species with the most widely spaced energy levels should have the fewest resonances in their cross sections and the most rapid fall off in cross section because of large energy gaps. This expectation has been confirmed in the studies reported above.

2. Technical developments: In order to experimentally pursue our goal of developing a collisional spectroscopy, we have built two new systems which allow us study collisions over the entire energy range represented by 1K to 1000 K. Specifically, by isolating the collisionally cooled cell from the helium bath, it is possible to do collisionally cooled spectroscopy above the 4 K boiling point of liquid helium. In addition to providing access to the region which represents the transition between phenomena best described quantum mechanically to the more semi-classical region, it allows us to study the collisions with hydrogen.

We have now begun to computationally investigate the relationship between cross sections predicted on the basis of *ab initio* interaction potentials and our experimental results. This is computationally very intensive. Accordingly, we have installed the MOLSCAT routines first on the North Carolina Cray and subsequently on the Ohio Cray. In our first investigations, we attempted (via a numerical nonlinear least squares fit) to adjust the intermolecular parameter to improve agreement between experiment and theory. Although we were able to reduce the deviation by about 300%, it is clear that systematic effects still remain.

10

B. Energy Transfer in Polyatomic Molecules

Our work on rotational and fast vibrational energy transfer grew out of an initial interest in constructing models of the internal dynamics of FIR molecular lasers which would include modern ideas of molecular collision dynamics. Although the number of degrees of freedom in these molecular systems might at first glance seem discouraging, we used mm/submm spectroscopic diagnostic techniques to establish *experimentally* the existence of pools of levels which are in equilibrium with each other, thereby substantially simplifying the resulting models. Because of the very high sensitivity of mm/submm spectroscopy to nonthermal rotational populations, this could be done with considerable certainty. Examples of such thermal pools include the equilibria of the vibrational bending modes with the R/T temperature in the HCN discharge FIR laser; all of the J rotational levels within the unpumped K = 1 state of the $^{12}CH_3F$ OPFIR laser; all of the rotational leve' within the unpumped symmetry species of the $^{13}CH_3F$ OPFIR, etc.[25,7,26] This work has pro ded a quantitative model of the complex HCN discharge laser system as well as significant advances in our understanding of OPFIR lasers. Perhaps the most interesting practical result of this latter work was the demonstration and theoretical explanation of the operation of an OPFIR laser in a high pressure, small cavity regime that was previously thought to be forbidden on rather fundamental grounds.[27]

In the work described below time resolved mm/submm-ir double resonance methods were used to obtain the experimental data which formed the basis for the development of numerical models which characterize the molecular energy transfer. The experimental details as well as the results of these investigations have been published in a number of papers.[7,8,26,28]

1. Modeling: As a first goal in our modeling of FIR lasers and rotational and vibrational energy transfer, we have sought to build the kinds of energy transfer models discussed above that describe the systems in terms of a finite number of individual energy levels and thermal pools along with the energy transfer rates that connect them together. Although the resulting models are too complex for analytical solution, we have found that they may be implemented numerically on microcomputers using relatively simple techniques. In this form nonlinear least squares techniques are used to adjust the rates of the model to match the time resolved data obtained from our experiments. At a more fundamental level we have sought to obtain rates that can be related to fundamental physical processes.

Briefly, we have found it possible to account for a large and diversified body of experimental data by:

(a) Collecting all of the rotational states of the symmetry species which does not contain the pumped transition into a single thermal pool, with the relative population of the levels

11

within the pool determined by the Boltzmann factor and with the total population free to vary in the model.

(b) Collecting all of the rotational states of the symmetry species which does contain the pumped transition into a similar pool, with the exception of the J states in the pumped K manifold which can have an excess 'nonthermal' population.

(c) Within the pumped K manifold, the nonthermal populations of all J are treated as independent variables.

(d) Additional pools for population transfer to vibrational states are included as required by the magnitude of the vibrational excitation. Typically, for the time resolved mm/submmir measurements of energy transfer rates, only the v_6 is required in addition to the ground vibrational state and the directly pumped v_3.[7,8] However, for strongly pumped cw laser systems we have found that population transfer to many additional states profoundly affects the laser operation.[27]

The principal energy transfer processes among these pools and states are:

(a) The $\Delta J = n$, $\Delta K = 0$ processes; the fastest of which is the electric dipole allowed $\Delta J = 1$.

(b) The spin statistic allowed $\Delta K = 3n$ processes, which distribute any nonequilibrium population according to a thermal distribution to all other rotational states of the same symmetry.

(c) A vibrational swap processes which, for example, *effectively* transfer population between the A and E symmetry species of the pumped excited vibrational state.

(d) Fast vibrational processes (e. g. $v_3 \rightarrow v_6$ or $v_3 + v_3 \rightarrow 2v_3$).

(e) Vibrational relaxation resulting from wall collisions.

2. Results: During this period we have made significant progress toward our goal of being able to understand collision induced rotational energy transfer on the basis of fundamental molecular interactions rather than as a collection of unrelated and unexplained state-to-state constants. Specifically we have finished a rather complete study of CH_3F in which we have succeeded in explaining a large amount of varied experimental data in terms of only a few molecular parameters; and more importantly, have used this same approach to predict the behavior of observations in CH_3Cl.

First let us consider CH_3F briefly. In this species a large number of transitions in a variety of vibrational levels and two different isotopic species were observed. In the end all of this seemingly complex behavior could be explained in terms of: 1) the permanent electric dipole moment, 2) the vibrational transition moment, 3) a cross section for $\Delta K = 3n$ processes, and 4) two constants which describe the Statistical Power Gap (SPG) law. Of these, only 4) are truly adjustable parameters. The first two are well known from other types

12

of experiments (i. e. measurements of Stark effects), and the third is closely related to the geometric cross section of the molecule. We consider this to be a significant accomplishment. This work has been summarized and more details are given in a recent publication.[29] These results are summarized in the Table below.

Collision Induced Transition Rates in CH_3F

	$^{12}CH_3F$	$^{13}CH_3F$
Gas kinetic collision cross section	44[a]	44
Dipole-Dipole cross section at 300K		320 (30)
$\Delta K = 3n$ cross section at 120K	33.1 (16)	46.6 (20)
$\Delta K = 3n$ cross section at 150K	28.6 (78)	47.0 (31)
$\Delta K = 3n$ cross section at 200K	51.1 (97)	54.8 (74)
$\Delta K = 3n$ cross section at 250K	46.0 (42)	74.1 (59)
$\Delta K = 3n$ cross section at 300K		137 (5)
$\Delta K = 3n$ cross section at 350K		190 (40)
$\Delta K = 3n$ cross section at 400K		235 (50)
v_2 V-swap cross section at 120K	50.6 (10)	54.1 (1)
v_2 V-swap cross section at 150K	43.4 (20)	52.3 (15)
v_2 V-swap cross section at 200K	36.4 (21)	36.2 (10)
v_2 V-swap cross section at 250K	22.8 (9)	30.2 (16)
v_2 V-swap cross section at 300K	18.9 (15)	21.0 (21)
v_2 V-swap cross section at 350K	15.6 (10)	16.8 (28)
v_2 V-swap cross section at 400K	13.4 (35)	13.6 (19)
$\Delta J = 2$ weighted cross section at 300K	2.36 (51)	5.10 (45)
$\Delta J = 3$ weighted cross section at 300K	1.60 (6)	2.61 (40)
$\Delta J = 4$ weighted cross section at 300K	1.14 (7)	1.82 (43)
$\Delta J = 5$ weighted cross section at 300K	0.897 (31)	1.27 (20)
$\Delta J = 6$ weighted cross section at 300K	0.622 (36)	1.03 (6)
$\Delta J = 7$ weighted cross section at 300K	0.510 (20)	
$\Delta J = 8$ weighted cross section at 300K	0.489 (25)	
$\Delta J = 9$ weighted cross section at 300K	0.438 (20)	
$\Delta J = 10$ weighted cross section at 300K	0.418 (15)	
a from SPG law at 300K ($Å^2$)	0.221 (6)	0.221 (6)
γ from SPG law at 300K	1.22 (3)	1.22 (3)
c for IOS cross sections at 300K ($Å^2$)	287 (68)	287 (68)
γ for IOS cross sections at 300K	1.30 (5)	1.30 (5)

[a] All cross sections are in $Å^2$.

More recently we have studied CH_3Cl. One of the foundations of our understanding of energy transfer in CH_3F was our obervation of thermal pools which could be used to describe the behavior of a large number of states of differing K, but of the same symmetry species. Our initial observations of this system appeared to show a much more complicated system, with widely varying rates associated with different K states. While this is a more 'interesting' result, we were concerned that our achievement of a simple physics which underlied the CH_3F system would not be possible. However, more careful consideration of the observations showed that exactly the same model could be applied to CH_3Cl if the CH_3F model were extended to allow an initial rotational temperature other than 300 K. In retrospect, this is not an unexpected result because of the wider range of levels pumped in CH_3Cl and because its lower B value would require larger changes in quantum number for

the collisions to redistribute the population according to the translational temperature (300 K). Moreover, we were extremely pleased that the value of the $\Delta K = 3n$ constant which results from this analysis is essentially identical (at constant collision velocity) as that observed in CH_3F. Additionally, the two constants which describe the SPG law are similar (they should not be exactly the same due to the differing geometry and molecular velocity) between the two species.

C. Millimeter and Submillimeter Technology

In order to support our experimental activity and to contribute to the mm/submm community our laboratory undertakes technology development as a part of its research program. Here we will briefly mention two effort in which we participate in collaboration with other research efforts in the laboratory.

We have had a long standing interest in the interaction between short (pico/femtosecond) pulses and the generation of mm/submm waves. In collaboration with John Swartz and Bob Guenther we are developing two techniques. The first uses a colliding pulse (soon to be replaced with a Ti:Sapphire laser) laser to produce femtosecond pulses of light which will in turn switch a semiconductor transmission line to generate mm/submm waves. We are especially interested in high resolution applications, based on signal processing of the demodulated optical pulse train. This work is based on our earlier work with a synchpump picosecond laser.[30] The second is a parasitic experiment on the bunched electron beam which drives the MKIII FEL. In this experiment significant amount of >100 GHz pulsed radiation has been produced. Here, we are both interested in the generation of radiation and the analysis of the mm/submm radiation produced as a diagnostic of the electron bunching parameters of the FEL.

In collaboration with Ron Jones and Jo Dutta of NCCU we have a related experiment which uses a microwave gun to produce an ~1 MeV beam of picosecond bunched electrons. In this dedicated experiment, interaction devices specifically designed to optimize desirable microwave source parameters (short pulses, high power, narrow bandwidth, etc.) have been designed and the first devices fabricated.

Participating Scientific Personnel

1. Frank C. De Lucia, Professor of Physics

2. Eric Herbrt, Professor of Physics

3. Paul Helminger, Professor of Physics, University of South Alabama

4. D. D. Skatrud, Associant Professor (adjunct)

5. B. D. Guenther, Professor (adjunct)

6. John Swartz, Graduate Student

7. S. H. Shostak, Research Associate

8. W. L. Ebenstein, Research Associate

9. Tom Goyette, Research Associate

10. Dan Wiley, Graduate Student

11. Todd Anderson, Graduate Student

12. Henry Everitt, Graduate Student

LIST OF REFEREED PUBLICATIONS

"Very low temperature spectroscopy: The pressure broadening coefficients for CO - He between 4.3 and 1.7 K," D. R. Willey, R. L. Crownover, D. N. Bittner, and F. C. De Lucia, J. Chem. Phys. 89, 1923 (1988).

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